Review for: “Spatial and temporal CCN variations in convection permitting aerosol microphysics simulations in an idealised marine tropical domain”

Summary and recommendation:

This article employs a convection-permitting resolution model to assess the contribution of spatial and temporal variations in aerosol properties for the case of a convective tropical marine boundary layer to CCN variability across a domain the size of a GCM grid box. The model is setup in a simplified idealised configuration in which the radiation scheme was turned off and CCN concentrations do not feed through to the cloud microphysics. Subsequently, the current setup ignores feedbacks associated with aerosol-radiative and aerosol-cloud microphysical interactions that may impact the simulated aerosol field in the model. The authors find that the simulated CCN concentrations can vary significantly over the domain, more than a factor of 8 during strongly convective conditions. They assess the contribution of dynamical, chemical and microphysical processes to this high variability in CCN and attribute it to increased sea salt/DMS emissions when spatial and temporal wind speed fluctuations become resolved at this convection-permitting resolution, increasing peak wind-speeds. This is an interesting finding as current GCMs cannot explicitly resolve sub-grid scale variability in wind speeds. Such modelling frameworks are required to elicit the impact of spatial/temporal resolution in GCMs on the representation of aerosol-cloud interactions. Therefore, I recommend publication of this article ACP once the following revisions have been addressed:

We thank the reviewer for their constructive review and address each of the specific comments raised below with our responses coloured red, and, where changes to the manuscript have been made, these are highlighted in the track-changes version of the document provided.

General comments:

- The modelling framework developed is described as ground-breaking. One of the key advantages of the model is in the use of a unified modelling (UM) framework to investigate the dependence of parameters involved in aerosol-cloud interactions on model resolution. However, this strength has not been captivated upon in this study. There is a lack of evaluation of the impact of the increased resolution in the model on the parameters of interest. A comparison of the domain averaged parameter values presented in the study to the same parameters simulated by the GCM would be highly beneficial and greatly strengthen the conclusions presented. Does the observed sub-grid scale variability in CCN impact the average CCN concentration across the domain compared to a GCM? This comparison should be provided before publication in ACP.

In our Figures 8 and 9 we specifically assess the CCN variability in the model, presenting the full pdf of the distribution in the vertical and at different points of the simulation across varying wind speed conditions. We agree strongly that comparing this CCN variability against that found from a lower resolution simulation with the same GCM would be a valuable comparison to make. Indeed the capability to nest down from global simulations with parameterized convection to limited-area domains runs at convection permitting resolution will enable this to be quantified in future studies with this Unified Model (UM) framework. However, the simulations here are with an idealized configuration of the UM whereby the solar variations

across a daily cycle are de-activated. We therefore feel comparing CCN variability across different horizontal resolution would best be reserved for a future study with the nested UM.

- CCN represent the aerosol particles that can form cloud droplets under reasonable atmospheric supersaturations. Accordingly, CCN concentrations always refer to a specific supersaturation, for example, CCN (0.1%) or CCN (0.5%) and one should be careful when comparing CCN concentrations measured or simulated at different supersaturations. What supersaturation was used throughout the article for the CCN concentrations presented?

Yes, we should have specified that, as in Mann et al. (2012), CCN concentrations are calculated here as soluble particles with dry diameter larger than 50nm, which corresponds to a supersaturation of 0.35%, calculated by Kohler theory for a pure sulphuric acid solution droplet. The marine aerosol review article by O'Dowd et al. (1997) refers to this threshold size as a good representative for activating nuclei. The revised manuscript now gives this definition for CCN at first use in the results section, referring to the specific threshold size and supersaturation used in the calculations.

The variability in CCN concentrations reported in this idealised configuration has been shown to be strongly dependant on variability in wind speed across the domain. This is unsurprising considering the strong wind-speed dependence of the sea spray emission parameterisation employed. Accordingly more discussion is required as to the sensitivity of the results presented on the choice of sea spray emission parameterisation with regard to the following:

- As the findings presented are strongly linked to the simulated wind speed field across the domain some discussion is required as to how accurate the simulated wind field and convective perturbation is compared to the real world. Also, is the aerosol, thus, CCN variability simulated expected compared to observations? Please discuss in relation to the footprint of flux measurements performed to measure sea-salt emissions in the marine environment and associated variability observed from these measurement campaigns.

We consider the model wind speed field across the domain, as simulated by the atmospheric dynamics in the MetUM, to be, for this type of model, highly realistic since the convection is explicitly resolved (no convection parameterization is required). The reviewer refers to the temporal footprint of flux measurements used to measure sea-salt emission. As we discuss in the reply to their later comment, that's an interesting point in relation to the time-window for the flux measurements used in deriving and evaluating the sea-spray source function flux used in our study. We consider this to be part of the future analysis in terms of applying the nested UM framework to assess the CCN variability in simulations with different spatial resolution.

- Numerous sea-salt emission parameterisations exist, derived from a variety of in-situ measurement campaigns and laboratory experiments. How does the chosen parameterisations wind-speed dependence compare to the range of parameterisations in the literature, e.g. Fig. 5 Salter et al., 2015? How might a different parameterisation alter the high variability in CCN across the domain found?

As we explain in the text, sea-salt is emitted according to the Gong (2003) sea-spray emission parameterization, as applied in our global model simulations (e.g. Mann et al., 2010). The Gong parameterization is based on the Monahan et al. (1986) sea-spray source function with a parameterization to better capture emissions of ultra-fine sea-spray. We have set the theta parameter controlling these ultra-fine sea-sprays to 30 in these simulations, as applied in Gong (2003). As in our reply to Reviewer 1, the Gong-Monahan emissions flux lie within the mid-range of other parameterizations in the literature (see e.g. Figure 5 of Salter et al., 2015),

so, in our view, it is reasonable to expect our results to be robust, with this sea-spray emissions parameterization particularly designed to capture the emitted ultra-fine sea spray.

• The onset of wave breaking is important for sea spray aerosol formation. It is generally recognized that the whitecap fraction and therefore sea spray aerosol production is zero for wind speeds less than ~ 3 m s^{-1} (Blanchard, 1963; Monahan, 1971). The implication of this with respect to the findings requires discussion, for example, what is the contribution of the total CCN variability simulated between 0-3 m s^{-1}? At what wind speeds does the CCN concentration begin to increase sharply, is there a threshold value?

There is no threshold velocity in the sea-spray source function, but, since the Gong parameterization is based on Monahan et al. (1986), it applies the emissions flux to be proportional to the 10m wind speed to the power 3.41, so the increase is quite steep as wind speed increases. Similarly, at the low windspeeds range mentioned (0-3 m/s) emissions fluxes will be low, and the non-linear dependence of the sea-spray emissions is one reason why this additional CCN variability becomes higher at this higher spatial resolution. We point out in the Introduction (page 3 lines 6-8) and feel this is explored sufficiently with the current text.

• Discussion is required on the applicability of the chosen parameterisation of the resolution of the model (1Km) and time-step. Typically sea salt emission parameterisations are applicable to certain footprints, and parameterisations developed from in-situ observations are dependent on the memory of the wave field (a rising sea will result in a different emission profile than a falling sea). In addition parameterisations are developed using longer time windows for averaging for flux measurements compared to the model time-step employed. Is the sea spray source function being applied in the model at this temporal/spatial resolution in the way it was designed?

As we state in the manuscript (page 5, lines 4-5), emissions are calculated (and enacted) every timestep of the simulation, which is 30 seconds at this high spatial resolution. Although the reviewer is absolutely correct to point out that wave state of the sea surface affects emitted sea-spray (e.g. Grythe et al., 2014, ACP), in our simulations with the Gong (2003) parameterization, these affects are not included. To address the reviewer comments we added the following to the revised manuscript:
"Other influences such as changes in sea surface wave state will also influence sea spray emissions (e.g. Grythe et al., 2014), but these effects are not resolved in this study. The Gong-Monahan parameterization used here is based on sea spray flux measurements made over a longer time period than the model timestep (30s), and observating capabilities now include eddy covariance sea-spray flux measurements (e.g. Norris et al., 2012), we expect our approach will resolve the dominant sources of sea spray emissions flux variability."

Minor comments:

• Section 2: A figure of the modified sea-spray source function used in the study would be beneficial here, especially for experimentalists.

We feel it is sufficient to reference the Gong (2003) paper. It's an established parameterization and was recommended for models to use in the AeroCom phase 1 co-ordinated experiment.

• Section 2: For a modelling framework described as ground-breaking the model description is relatively sparse, for instance, how is hygroscopic growth parameterised in the model? This

will affect the evolution of the aerosol field across the domain. Please provide a more detailed description of the aerosol microphysics scheme.

The ground-breaking aspect of this study is the ability to use a new numerical framework that is based on a coupling between the UKCA detailed aerosol module and the MetUM model at very high scale.

Regarding the hygroscopic growth, this aerosol process is parameterized thanks to the ZSR method (Zadanovksii, 1948; Stokes and Robinson, 1966) using data from Jacobson et al. (1996) to calculate the binary electrolyte molalities. The complete description of the hygroscopic growth parameterization as well as the description of all the others aerosol processes of the model are described in details in Mann et al. (2010).

We modified the manuscript in order to inform the reader that a complete description of the different aerosol processes is provided in this paper.

- Section 2.2: It is widely known 1-moment cloud microphysics schemes introduce errors compared to 2 or3 moment schemes. Some justification of this choice is required, was it due to computational restraints?

Yes, in this study a single-moment microphysics scheme is used as it is the only one currently available in the latest version of the Unified Model at that time. We clarified the description of the microphysics scheme in the manuscript.

- Section 3.2, line 8: "Aitken mode are almost exclusively secondary in nature": Please reword, this is two strong, studies exist which show emission of sea spray in this size regime, e.g. Salter et al., 2015.

We clarified the manuscript.

- Section 4, line 25. "comprising two elements": reword.

We clarified the manuscript.

- Fig.4: Why do the error bars in DMS & SO2/H2SO4 not correspond? Some discussion on expected oxidation timescales required, why is there no offset between H2SO4 & DMS observed?

We explain in the text that the steps involved for the SO2 and H2SO4 to be produced following oxidation in the atmosphere and we do not understand the reviewer's point here. We feel the existing text is sufficient here to explain what is shown in the Figure.

- Recent studies have probed the dependence of aerosol processes on model resolution, for instance Weigum et al., 2016. This should be referenced.

We added this reference in the introduction section.

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